

PATENT ABSTRACTS OF JAPAN

(11)Publication number : 2002-012650

(43)Date of publication of application : 15.01.2002

(51)Int.Cl.

C08G 59/42
H05K 1/03

(21)Application number : 2000-198517 (71)Applicant : DAINIPPON INK & CHEM INC

(22)Date of filing : 30.06.2000 (72)Inventor : USAMI SUKEAKI
DEMURA SATOSHI
TAKAHASHI KATSUJI

(54) EPOXY RESIN COMPOSITION FOR LOW-DIELECTRIC MATERIAL

(57)Abstract:

PROBLEM TO BE SOLVED: To provide an epoxy resin composition for low-dielectric materials having excellent adhesion, solder resistance and curability and assuming a low permittivity and/or a low dielectric dissipation factor and a cured product obtained therefrom.

SOLUTION: This epoxy resin composition for the low-dielectric materials consists essentially of (A) an epoxy resin having ≥ 2 epoxy groups in one molecule, (B) an aromatic compound obtained from a compound having a polyvalent carboxylic acid and an aromatic compound having a phenolic hydroxyl group and having ≥ 2 active ester groups in one molecule and (C) a reaction accelerator.

LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of rejection]

[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

AN 2002:35832 CAPLUS
DN 136:86946
ED Entered STN: 15 Jan 2002
TI Epoxy resin compositions and their low dielectric cured materials
IN Usami, Hiroaki; Demura, Satoru; Takahashi, Katsuji
PA Dainippon Ink and Chemicals, Inc., Japan
SO Jpn. Kokai Tokkyo Koho, 14 pp.
CODEN: JKXXAF

DT Patent
LA Japanese
IC ICM C08G059-42
ICS H05K001-03
CC 38-3 (Plastics Fabrication and Uses)
Section cross-reference(s): 37, 76

FAN.CNT 1

	PATENT NO.	KIND	DATE	APPLICATION NO.	DATE
PI	JP 2002012650	A2	20020115	JP 2000-198517	20000630 <--
PRAI	JP 2000-198517		20000630		

CLASS

PATENT NO.	CLASS	PATENT FAMILY CLASSIFICATION CODES	
JP 2002012650	ICM	C08G059-42	
	ICS	H05K001-03	
	IPCI	C08G0059-42 [ICM,7]; H05K0001-03 [ICS,7]	<--

AB The compns. contain (A) epoxy resins bearing ≥ 2 epoxy groups, (B) aromatic compds. bearing ≥ 2 active ester groups prepared from polycarboxylic acids and aromatic compds. bearing phenolic OH, and (C) reaction accelerators. The compns. show excellent adhesion, solder resistance, curability, low permittivity and/or low dielec. loss and are useful for printed circuit boards, laminates, elec. insulators, adhesives, etc. Thus, 0.2 mol isophthalic acid, β -naphthol 0.4, and Ac2O 0.80 mol were treated at 145° for acetylation, followed with deacetylation and esterification while heating to give a 62.5 g of an active ester group-containing aromatic compound with esterification degree

96.9%,
100 parts of which was mixed with an epoxy resin (Epiclon N 660) 100, an accelerator (2E4Mz) 0.60, and NMP 400 parts to give a varnish, and cured in an Al container by heating to give a test piece having Tg 135°, permittivity 3.18, 3.15, and 3.08 and $\tan\delta$ 0.0051, 0.0068, and 0.0091 at 1 MHz, 30 MHz, and 1 GHz, resp., and linear expansion coefficient 50/°C + 106. Peeling strength for Cu foil from its Cu clad laminate had 1.2 kN/m.

ST epoxy resin hardener arom active ester; polycarboxylic acid phenolic hydroxy active ester

IT Epoxy resins, uses
RL: IMF (Industrial manufacture); PRP (Properties); TEM (Technical or engineered material use); PREP (Preparation); USES (Uses)
(crosslinked; low dielec. epoxy resins crosslinked with active esters of aromatic compds.)

IT Crosslinking agents
(epoxy resin compns. containing active esters of aromatic compds. and their low dielec. cured materials)

IT Phenolic resins, uses
RL: POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); USES (Uses)
(epoxy, novolak, cresolic; epoxy resin compns. containing active esters of aromatic compds. and their low dielec. cured materials)

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RL: POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); USES (Uses)
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RL: POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); USES (Uses)

(phenolic, novolak, cresolic; epoxy resin compns. containing active esters of aromatic compds. and their low dielec. cured materials)

IT Epoxy resins, uses

RL: POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); USES (Uses)

(phenolic, novolak; epoxy resin compns. containing active esters of aromatic compds. and their low dielec. cured materials)

IT 387336-28-9P, Epiclon N 660- α -naphthol isophthalate copolymer
387336-29-0P, Epiclon N 660- β -naphthol isophthalate copolymer
387336-30-3P, Epiclon N 660- α -naphthol terephthalate copolymer
387336-31-4P, Epiclon N 770- α -naphthol isophthalate copolymer
387336-32-5P, Epiclon N 770- β -naphthol trimellitate copolymer
387336-33-6P, Epiclon N 660- β -naphthol adipate copolymer

RL: IMF (Industrial manufacture); PRP (Properties); TEM (Technical or engineered material use); PREP (Preparation); USES (Uses)

(epoxy resin compns. containing active esters of aromatic compds. and their low dielec. cured materials)

IT 95916-94-2 $\frac{1}{2}$, Epiclon N 660 99241-45-9 $\frac{1}{2}$, Epiclon N 770

RL: POF (Polymer in formulation); PRP (Properties); TEM (Technical or engineered material use); USES (Uses)

(epoxy resin compns. containing active esters of aromatic compds. and their low dielec. cured materials)

(IT) 387336-23-4P $\frac{1}{2}$, α -Naphthol terephthalate 387336-24-5P $\frac{1}{2}$,
 α -Naphthol isophthalate 387336-25-6P $\frac{1}{2}$, β -Naphthol isophthalate
387336-26-7P $\frac{1}{2}$, β -Naphthol trimellitate 387336-27-8P $\frac{1}{2}$, β -Naphthol
adipate

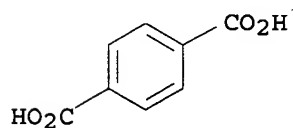
RL: IMF (Industrial manufacture); MOA (Modifier or additive use); TEM (Technical or engineered material use); PREP (Preparation); USES (Uses)

(hardener; epoxy resin compns. containing active esters of aromatic compds. and their low dielec. cured materials)

RN 387336-23-4 REGISTRY
ED Entered STN: 28 Jan 2002
CN 1,4-Benzenedicarboxylic acid, 1-naphthalenyl ester (9CI) (CA INDEX NAME)
OTHER NAMES:
CN α -Naphthol terephthalate
MF C10 H8 O . x C8 H6 O4
CI COM
SR CA
LC STN Files: CA, CAPLUS

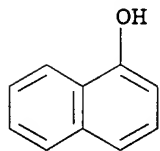
CM 1

CRN 100-21-0
CMF C8 H6 O4



CM 2

CRN 90-15-3
CMF C10 H8 O

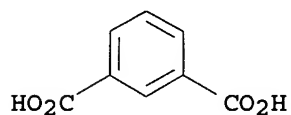


1 REFERENCES IN FILE CA (1907 TO DATE)
1 REFERENCES IN FILE CAPLUS (1907 TO DATE)

RN 387336-24-5 REGISTRY
ED Entered STN: 28 Jan 2002
CN 1,3-Benzenedicarboxylic acid, 1-naphthalenyl ester (9CI) (CA INDEX NAME)
OTHER NAMES:
CN α -Naphthol isophthalate
MF C10 H8 O . x C8 H6 O4
CI COM
SR CA
LC STN Files: CA, CAPLUS

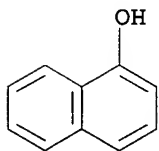
CM 1

CRN 121-91-5
CMF C8 H6 O4



CM 2

CRN 90-15-3
CMF C10 H8 O

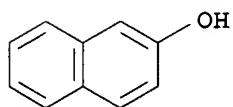


3 REFERENCES IN FILE CA (1907 TO DATE)
3 REFERENCES IN FILE CAPLUS (1907 TO DATE)

RN 387336-25-6 REGISTRY
ED Entered STN: 28 Jan 2002
CN 1,3-Benzenedicarboxylic acid, 2-naphthalenyl ester (9CI) (CA INDEX NAME)
OTHER NAMES:
CN β -Naphthol isophthalate
MF C10 H8 O . x C8 H6 O4
CI COM
SR CA
LC STN Files: CA, CAPLUS

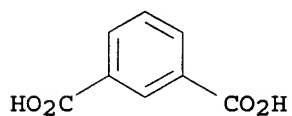
CM 1

CRN 135-19-3
CMF C10 H8 O



CM 2

CRN 121-91-5
CMF C8 H6 O4



1 REFERENCES IN FILE CA (1907 TO DATE)
1 REFERENCES IN FILE CAPLUS (1907 TO DATE)

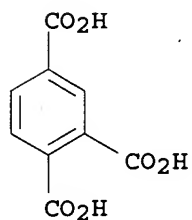
RN 387336-26-7 REGISTRY
ED Entered STN: 28 Jan 2002
CN 1,2,4-Benzenetricarboxylic acid, 2-naphthalenyl ester (9CI) (CA INDEX NAME)

OTHER NAMES:

CN β -Naphthol trimellitate
MF C10 H8 O . x C9 H6 O6
CI COM
SR CA
LC STN Files: CA, CAPLUS

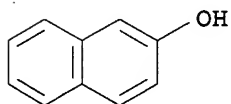
CM 1

CRN 528-44-9
CMF C9 H6 O6



CM 2

CRN 135-19-3
CMF C10 H8 O

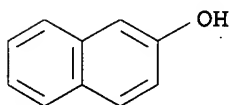


1 REFERENCES IN FILE CA (1907 TO DATE)
1 REFERENCES IN FILE CAPLUS (1907 TO DATE)

RN 387336-27-8 REGISTRY
ED Entered STN: 28 Jan 2002
CN Hexanedioic acid, 2-naphthalenyl ester (9CI) (CA INDEX NAME)
OTHER NAMES:
CN β -Naphthol adipate
MF C10 H8 O . x C6 H10 O4
CI COM
SR CA
LC STN Files: CA, CAPLUS

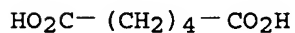
CM 1

CRN 135-19-3
CMF C10 H8 O



CM 2

CRN 124-04-9
CMF C6 H10 O4



1 REFERENCES IN FILE CA (1907 TO DATE)
1 REFERENCES IN FILE CAPLUS (1907 TO DATE)

DERWENT-ACC-NO: 2002-540890

DERWENT-WEEK: 200264

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TITLE: Epoxy resin composition for low dielectric material,
e.g., substrate of integrated circuit, contains active
ester group having aromatic compound as curing agent

PATENT-ASSIGNEE: DAINIPPON INK & CHEM INC[DNIN]

PRIORITY-DATA: 2000JP-0198517 (June 30, 2000)

PATENT-FAMILY:

PUB-NO	PUB-DATE	LANGUAGE	PAGES	MAINIPC
<u>JP 2002012650 A</u>	January 15, 2002	N/A	014	C08G 059/42

APPLICATION-DATA:

PUB-NO	APPL-DESCRIPTOR	APPL-NO	APPL-DATE
JP2002012650A	N/A	2000JP0198517	June 30, 2000

INT-CL (IPC): C08G059/42, H05K001/03

ABSTRACTED-PUB-NO: JP2002012650A

BASIC-ABSTRACT:

NOVELTY - An epoxy resin composition for a low dielectric material contains:

(A) epoxy resin;

(B) aromatic compound which has at least two active ester groups and is
produced from multi-valent carboxylic acid having compound and a phenolic
hydroxyl group having aromatic compound; and

(C) reaction promoter.

USE - The epoxy resin composition is useful for a low dielectric material
having dielectric constant of 3.5 or less and dielectric tangent of 0.01 or
less or a low-dielectric substrate for a printed circuit board, electrical
insulating resin or adhesive resin.

ADVANTAGE - The epoxy resin composition has good adhesiveness, soldering
resistance, curing properties, low dielectric constant and/or low dielectric
tangent after curing.

CHOSEN-DRAWING: Dwg.0/0

TITLE-TERMS: EPOXY RESIN COMPOSITION LOW DIELECTRIC MATERIAL SUBSTRATE
INTEGRATE CIRCUIT CONTAIN ACTIVE ESTER GROUP AROMATIC COMPOUND CURE
AGENT

DERWENT-CLASS: A21 A85 G03 L03 U11 V04 X12

CPI-CODES: A08-D; A08-D02; A09-A03; A12-E07C; G03-B02E2; L03-A03A; L03-H04E1;

EPI-CODES: U11-A07; V04-R07L; X12-E02B;

ENHANCED-POLYMER-INDEXING:

Polymer Index [1.1]

018 ; P0464*R D01 D22 D42 F47

Polymer Index [1.2]

018 ; ND01 ; ND04 ; B9999 B3203*R B3190 ; Q9999 Q7476 Q7330 ; B9999

* NOTICES *

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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] the thing concerning [this invention] an epoxy resin constituent and its hardened material -- it is -- still more detailed dielectric characteristics useful as the resin for printed circuit boards, the resin for laminates, the resin for electric insulation, and various resin for adhesives especially a low dielectric constant, and a low dielectric dissipation factor -- presenting -- in addition -- and it is related with the epoxy resin constituent excellent also in an adhesive property, solder-proof nature, and hardenability and its hardened material and the low dielectric ingredient that used this constituent for the list, for example, a substrate, a laminate, etc.

[0002]

[Description of the Prior Art] From the former, the epoxy resin excellent in the electrical property, the mechanical characteristic, the adhesive property, etc. has been widely used for the laminate, the printed circuit board, etc. While recent years come, a highly informative society is greeted and amount of information becomes huge, the densification of much more printed-circuit board and the Kota stratification progress, high heat-resisting material is called for, and on the other hand, improvement in the speed of the signal in various electronic equipment and a high-frequency communication link become indispensable, improvement in a transmission characteristic is called for, and a low insulating material of a dielectric constant and a dielectric dissipation factor is desired. Especially for a communication link, reduction of a dielectric dissipation factor becomes important.

[0003] However, in the usual epoxy resin, in the reaction with that curing agent, since the ring breakage of an epoxy group happened, the polar high hydroxyl group was able to generate in connection with it, and a dielectric constant and a dielectric dissipation factor were not able to be reduced for this hydroxyl group.

[0004] The epoxy resin hardened material using the compound which contains an activity ester group as a curing agent of an epoxy resin is proposed as a low dielectric constant ingredient as shown in JP,11-71499,A, JP,11-71500,A, and JP,11-130939,A in recent years. Moreover, the modified epoxy resin which used the activity ester group content aromatic compound as a modifier, and its manufacture approach are indicated by JP,7-292067,A, and it is proposed as a low dielectric constant ingredient.

[0005] However, the activity ester content compound which is the modifier used for the compound or modified epoxy resin containing the activity ester group which is the curing agent used in these esterifies the phenolic hydroxyl group of a polyhydric-phenol compound with monocarboxylic acid, and, as for the compound which has the multiple-valued carboxylic acid of the invention in this application, and the aromatic compound which has two or more activity ester groups in 1 molecule obtained from the aromatic compound which has a phenolic hydroxyl group, configurations completely differ. Moreover, since invention concerning JP,7-292067,A contains the curing agent which is not in the invention in this application as an indispensable component, it differs from the invention in this application. Furthermore, when an ester part is cut, in order that a phenol component may generate the activity ester group content aromatic compound which esterified the phenolic hydroxyl group of a

polyhydric-phenol compound with monocarboxylic acid as a free acid, about [that insulation becomes low], a dielectric constant, and a dielectric dissipation factor also become high.

[0006] Moreover, reference is not made at all by JP,5-51517,A, JP,5-263058,A, and JP,5-271637,A about a dielectric, and the polyester in the resin hardened material which heat-hardened the thermosetting resin constituent which uses as an indispensable component the epoxy resin shown in this official report and polyester does not necessarily show a low dielectric.

[0007] Moreover, although the activity ester which consists of a multiple-valued carboxylic acid was used in the activity ester shown in JP,11-349666,A, JP,6-293823,A, JP,63-57614,A, JP,63-30451,A, JP,62-212416,A, and JP,8-120057,A, it was not understood at all whether neither would be mentioned at all by the dielectric of an epoxy resin, but would have a low dielectric.

[0008]

[Problem(s) to be Solved by the Invention] The technical problem which this invention tends to solve makes it a technical problem to offer the epoxy resin constituent which is excellent in an adhesive property, solder-proof nature, and hardenability, and gives a dielectric constant and/or the low epoxy resin hardened material of a dielectric dissipation factor.

[0009]

[Means for Solving the Problem] Without producing a free acid by using an activity ester group content aromatic compound as a curing agent of an epoxy resin, as a result of repeating research wholeheartedly that the technical problem mentioned above should be solved, this invention person finds out that the dielectric which does not make a polar high hydroxyl group generate at the time of hardening was excellent, namely, the epoxy resin constituent which gives the epoxy resin hardened material of a low dielectric constant and/or a low dielectric dissipation factor can be obtained, and came to complete this invention.

[0010] That is, this invention is [0011]. (1) The epoxy resin (A) which has two or more epoxy groups in 1 molecule, the aromatic compound (B) which has two or more activity ester groups in 1 molecule obtained from the compound which has a multiple-valued carboxylic acid, and the aromatic compound which has a phenolic hydroxyl group, the epoxy resin constituent for low dielectric ingredients which

uses a reaction accelerator (C) as an indispensable component, [0012] (2) The epoxy resin constituent for low dielectric ingredients given in (1) to which a low dielectric is characterized by being 3.5 or less dielectric constant, [0013] (3) (1) to which a low dielectric is characterized by being 0.01 or less dielectric dissipation factor or the epoxy resin constituent for low dielectric ingredients given in (2),

[0014] (4) the activity ester group content aromatic compound (B) to the epoxy resin (A) which has two or more epoxy groups in 1 molecule -- comparatively -- the weight per epoxy equivalent of this epoxy resin (A) -- receiving -- the ester equivalent of this aromatic compound (B) -- 0.3-1.5Eq -- it is -- [0015]

The epoxy resin constituent for low dielectric ingredients given in any 1 term of (1) - (3) to which a reaction accelerator (C) is characterized by being the 0.005 - 10.0 weight section to a total of 100 weight sections of an epoxy resin (A) and an activity ester group content aromatic compound (B), [0016] (5)

The aromatic compound (B) which has two or more activity ester groups in 1 molecule obtained from the compound which has a multiple-valued carboxylic acid, and the aromatic compound which has a phenolic hydroxyl group is [0017]. a multiple valued -- a carboxylic acid -- having -- an aromatic

compound -- a phenolic hydroxyl group -- having -- an aromatic compound -- from -- obtaining --

having -- one -- a molecule -- inside -- two -- a piece -- more than -- activity -- an ester group -- having -

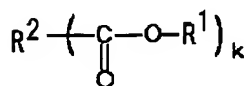
- all -- an aromatic compound (B') -- it is -- things -- the description -- ** -- carrying out -- (-- one --) - (-

- four --) -- some -- one -- a term -- a publication -- low -- a dielectric -- an ingredient -- ** -- an epoxy

resin -- a constituent -- [-- 0018 --] (6) The epoxy resin constituent for low dielectric ingredients given in any 1 term of (1) - (4) characterized by the aromatic compound (B) which has two or more activity ester groups in 1 molecule obtained from the compound which has a multiple-valued carboxylic acid, and the aromatic compound which has a phenolic hydroxyl group being an aromatic compound expressed with a general formula (1), [0019] General formula (1)

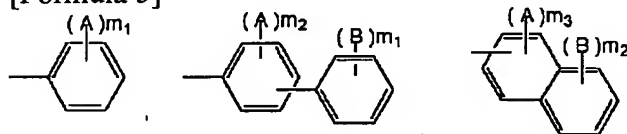
[0020]

[Formula 4]



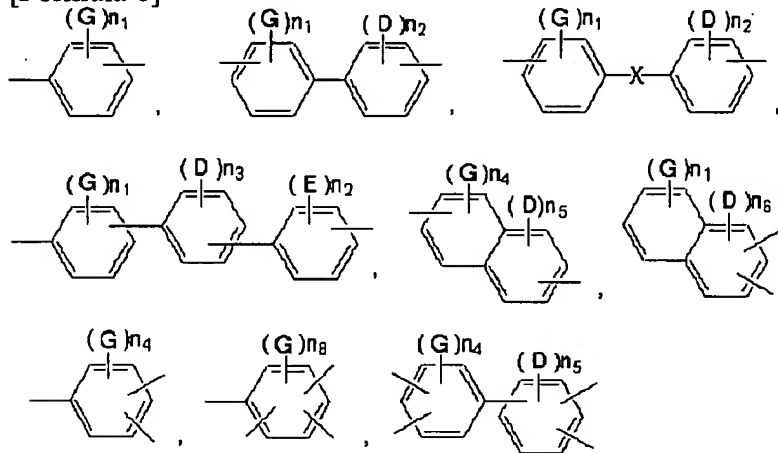
[0021] k is the integer of 2-4 among [type, and R1 is the three following formulas [0022].

[Formula 5]



[0023] (-- the inside of a formula, and A and B -- each -- the same -- or it differs, a halogen atom and an alkyl group are shown, and, as for 0-5, and m2, m1 shows 0-3, as for 0-4, and m3. D -- the aromatic series ring expressed with any one -- it is -- 0024] R2 is the nine following types [0025].

[Formula 6]



[0026] (-- the inside of a formula, and D, E and G -- each -- independently the same -- or -- differing -- a halogen atom and an alkyl group -- being shown -- X -- a sulfur atom, an oxygen atom, SO2, or CO -- being shown -- n1, n2, and n3 -- each -- independent -- 0-4, and n4 and n5 -- each -- 0-3, and n6 show 0-2 independently.) -- it is the aromatic series ring expressed with any one.]

[0027] (7) The epoxy resin constituent for low dielectric ingredients given in any 1 term of (1) - (6) characterized by the weight per epoxy equivalent of the epoxy resin (A) which has two or more epoxy groups in 1 molecule being 100-1000, [0028] (8) The epoxy resin constituent for low dielectric ingredients given in any 1 term of (1) - (7) characterized by reaction accelerators (C) being an imidazole compound, an organic phosphine compound, an organic phosphite compound, phosphonium salt, amines and its salt, a urea compound, bases, and one or more sorts of mixture chosen from the group which it becomes from the salt of crown ether, [0029] (9) (1) The low dielectric ingredient, [0030] which harden the epoxy resin constituent for low dielectric ingredients of a publication in any 1 term of - (8), and are obtained (10) The low dielectric substrate, [0031] which are characterized by having a metallic foil to one side or both sides of the film set to (9) from the low dielectric ingredient of a publication, or a sheet It provides.

[0032]

[Embodiment of the Invention] The epoxy resin constituent for low dielectric ingredients of this invention The low dielectric which it not only excels in an adhesive property, solder-proof nature, and hardenability, but was further excellent, Namely, the epoxy resin which has two or more epoxy groups in a specific presentation component, i.e., 1 molecule, in order it is important to present a low dielectric constant and/or a low dielectric dissipation factor and to satisfy these demand characteristics (A) (it is hereafter called above "an epoxy resin (A)" if needed.) The aromatic compound which has two or more activity ester groups in 1 molecule obtained from the compound which has a multiple-valued carboxylic

acid, and the aromatic compound which has a phenolic hydroxyl group (B) (hereafter above) it is called "an activity ester group content aromatic compound (B)" if needed. It is necessary to use the epoxy resin constituent for low dielectric ingredients which uses a reaction accelerator (C) as an indispensable component.

[0033] However, in this invention, a "low dielectric" means a low dielectric constant and/or a low dielectric dissipation factor, and the 100kHz - 10GHz dielectric constants in a room temperature are 2.0-3.2 still more preferably 3.2 or less preferably 3.5 or less, and it means in more detail that dielectric dissipation factors are 0.001-0.009 still more preferably 0.009 or less preferably 0.01 or less.

[0034] Moreover, in this invention, a "low dielectric ingredient" means the ingredient used for the application which needs a low dielectric, and the epoxy resin constituent for low dielectric ingredients means the epoxy resin constituent used for a low dielectric ingredient.

[0035] Generally, if thermal resistance goes up an epoxy resin hardened material, an adhesive property which is represented by copper foil Peel reinforcement will fall, but the good hardened material of the balance having a strong adhesive property is called for, maintaining thermal resistance, and the glass transition temperature whose epoxy resin constituent of this invention is a heat-resistant index gives the epoxy resin hardened material with which the copper foil Peel reinforcement which is 130 degrees C or more, and is an adhesive index is satisfied of 1.2 or more kN/m.

[0036] Weight per epoxy equivalent is 100-1000, the epoxy resin of low - inside molecular weight (300-5000) is mentioned, and the epoxy resin (A) which has two or more epoxy groups in 1 molecule of this invention is [0037]. Specifically For example, a cresol novolak, a phenol novolak, A naphthol denaturation novolak, bisphenol A, bisphenol - F, Tetrabromobisphenol A, a biphenyl mold epoxy resin, a triphenyl mold epoxy resin, The glycidyl ether mold epoxy resin of phenol systems, such as a tetraphenyl mold epoxy resin, The glycidyl ether mold epoxy resin of alcoholic systems, such as a polypropylene glycol and hydrogenation bisphenol A, The dicyclopentadiene mold epoxy resin containing a dicyclopentadiene frame, The glycidyl ester mold epoxy resin which used the naphthalene mold epoxy resin and hexahydro phthalic anhydride containing a naphthalene frame, dimer acid, etc. as the raw material, The glycidyl amine mold epoxy resin which used polyamine, such as diamino diphenylmethane, as the raw material, an alicyclic mold epoxy resin, [0038] Although bromination epoxy resins, those mixture, etc. are mentioned, in order to obtain good thermal resistance in it, one or more sorts of mixture of the epoxy resin chosen from a cresol novolak mold epoxy resin, a phenol novolak mold epoxy resin, a naphthol denaturation novolak mold epoxy resin, a bisphenol mold epoxy resin, a biphenyl mold epoxy resin, a triphenyl mold epoxy resin, a tetra-phenyl mold epoxy resin, a dicyclopentadiene mold epoxy resin, a naphthalene mold epoxy resin, and a bromination epoxy resin is desirable.

[0039] the compound (I) with which the aromatic compound (B) which has two or more activity ester groups in 1 molecule obtained from the compound which has the multiple-valued carboxylic acid of this invention, and the aromatic compound which has a phenolic hydroxyl group has at least two or more carboxylic acids in 1 molecule, and the aromatic compound (II) which has a phenolic hydroxyl group -- since -- it is the aromatic compound obtained and is the aromatic compound which has two or more ester groups in the molecule of this aromatic compound. Moreover, the straight chain and the dendritic macromolecule may also be contained. Moreover, if a compound (I) is a compound containing an aliphatic series chain, compatibility with an epoxy resin (A) can be made high, and thermal resistance can be made high if it is the compound which has an aromatic series ring.

[0040] Furthermore, in this invention, in order to give thermal resistance, it is desirable to use all the aromatic compounds (B') (above henceforth "an activity ester group content all aromatic compound (B)" if needed) that have two or more activity ester groups in 1 molecule obtained from the aromatic compound which has a multiple-valued carboxylic acid, and the aromatic compound which has a phenolic hydroxyl group as an activity ester group content aromatic compound (B) as a curing agent,

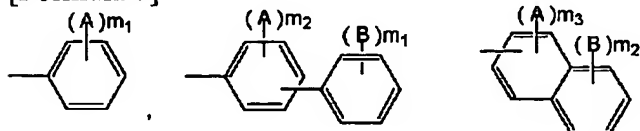
[0041] However, in this invention, all aromatic compounds mean the aromatic compound which does not have at all the aliphatic series chain with which direct coupling of the aromatic series ring contained in a compound is carried out to other aromatic series rings in one or more ester groups in the molecule

obtained from the aromatic compound which has a multiple-valued carboxylic acid, and the aromatic compound which has a phenolic hydroxyl group, and/or a molecule, and it combines them, and this compound can present high thermal resistance and a high low dielectric, i.e., a low dielectric constant, and/or a low dielectric dissipation factor. Moreover, you may be a straight chain and a dendritic macromolecule also about the case of these all aromatic compounds.

[0042] For example, all the aromatic polyester of the high heatproof mold of tetramethyl biphenol, isobutyl terephthalate, or the terephthalic-acid origin is also preferably used as an activity ester group content aromatic compound (B) of this invention.

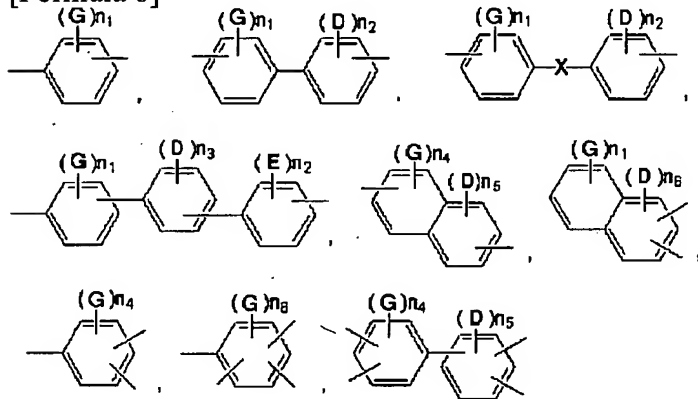
[0043] Here, as a soil skeleton of the aromatic series ring in [all / that was mentioned above] an aromatic compound (B'), they are the three following types [0044].

[Formula 7]



[0045] (-- A and B -- each -- the same -- or it differs, a halogen atom and an alkyl group are shown, and, as for 0-5, and m2, m1 shows 0-3, as for 0-4, and m3.) -- the univalent soil skeleton (hereafter referred to as "R1".) expressed with any one mentions -- having -- [0046] Or the following 9 types [0047]

[Formula 8]



[0048] the inside of a formula, and D, E and G -- each -- independently the same -- or -- differing -- a halogen atom -- an alkyl group -- being shown -- X -- a sulfur atom, an oxygen atom, SO₂, or CO -- being shown -- n1, n2, and n3 -- each -- independent -- 0-4, and n4 and n5 -- each -- 0-3, and n6 show 0-2 independently. The soil skeleton of many ** expressed with any one (it is hereafter referred to as "R2".) It is mentioned.

[0049] Moreover, as a halogen atom of A, B, D, E, or G, a fluorine atom, a chlorine atom, a bromine atom, and an iodine atom are mentioned, and a fluorine atom, a chlorine atom, and a bromine atom are raised preferably.

[0050] moreover, as an alkyl group of A, B, D, E, or G Although especially a carbon number is not limited, preferably A methyl group, an ethyl group, n-propyl group, An isopropyl group, n-butyl, sec-butyl, tert-butyl, n-pentyl radical, an isopentyl radical, a neopentyl radical, a tert-pentyl radical, n-hexyl group and a 4-methyl-2-pentyl radical -- it passes, a PUCHIRU radical, an octyl radical, a nonyl radical, a decyl group, an undecyl radical, the dodecyl, etc. are raised, and the alkyl group of carbon numbers 1-3 is raised preferably. X shows a sulfur atom, an oxygen atom, SO₂, or CO.

[0051] As for 0-5, and m2, m1 shows 0-3, as for 0-4, and m3. As for n1, n2, and n3, in 0-4, and n4 and n5, 0-3, and n6 show 0-2 independently respectively.

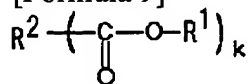
[0052] Furthermore, the ester compound with which the activity ester group content all aromatic compound (B') of this invention connected two or more R1 components and one or more R2 components by the direct ester bond is mentioned preferably.

[0053] Furthermore, all the aromatic compounds expressed with a general formula (1) are concretely mentioned more preferably as an activity ester group content all aromatic compound (B').

[0054] General formula (1)

[0055]

[Formula 9]



[0056] However, k is the integer of 2-4 among a formula, and R1 and R2 are as having already given the definition.

[0057] Since this invention can press down the free acid produced by hydrolysis as much as possible as a curing agent when it is used as an epoxy resin hardened material an activity ester group content aromatic compound (B) and by using an activity ester group content all aromatic compound (B') more preferably, it presents a low dielectric.

[0058] the case where the activity ester group content aromatic compound (B) of this invention requires liquid-like workability -- molecular weight 300-5000 -- it is 300-1000 preferably.

[0059] As an example of the synthesis method of the activity ester group content all aromatic compound (B) of this invention, a synthesis method with conventionally well-known an acetic-anhydride method, an interface method, a direct method, etc. is mentioned.

[0060] For example, by the acetic-anhydride method, after acetylating the aromatic compound which has a phenolic hydroxyl group with the acetic anhydride of an excess, the method of obtaining an activity ester group content aromatic compound is mentioned by performing the compound and decarboxylation reaction which have a multiple-valued carboxylic acid. In order to perform acetylation sufficient as an amount of acetic anhydrides, a phenolic hydroxyl group and more than equimolar are desirable.

[0061] By the interface method, the approach of contacting the organic phase containing the acid chloride of a multiple-valued carboxylic acid and the aqueous phase containing the aromatic compound containing a phenolic hydroxyl group, and obtaining an activity ester group content aromatic compound can be mentioned. As a solvent used for an organic phase, it is the solvent of nonaqueous solubility in which the acid chloride of a multiple-valued carboxylic acid is made to dissolve, for example, toluene, a hexane, etc. are desirable.

[0062] As an aliphatic series system carboxylic acid used in order to mainly increase compatibility with an epoxy resin, specifically as a compound used for the raw material of an activity ester group content aromatic compound (B) A malonic acid, a succinic acid, a glutaric acid, an adipic acid, a sebacic acid, a fumaric acid, A maleic acid, an itaconic acid, aconitic acid, tricarballic acid, 1, 2 and 3, 4? butane tetracarboxylic acid, 4? methyl 74? cyclohexene ?1, 2? dicarboxylic acid, 1, 2 and 3, 4? cyclopentane tetracarboxylic acid, The aliphatic series multiple-valued carboxylic acid or its anhydride, and acid chloride of saturation, such as 5?(2, 5? dioxo tetrahydro ?3? furanyl) ?3? methyl ? cyclohexene ?1 and 2? dicarboxylic acid anhydride, or partial saturation are mentioned. Moreover, as an aromatic series system carboxylic acid used in order to mainly increase thermal resistance, it is benzene dicarboxylic acid, such as naphthoic acids, such as benzoic acids, such as a benzoic acid, a methyl benzoic acid, a dimethyl benzoic acid, and a trimethyl benzoic acid, 1-naphthoic acid, and 2-naphthoic acid, a phthalic acid, isophthalic acid, and a terephthalic acid, or the acid anhydride of those and an acid chloride, trimellitic acid, and [0063]. Naphthalene dicarboxylic acid [, such as tetracarboxylic acid /, such as -BIENIREN tetracarboxylic acid, / or acid-anhydride / of those / 1, 4-naphthalene dicarboxylic acid, 2, 6-naphthalene dicarboxylic acid, 2, and 3-naphthalene dicarboxylic acid,] or acid-anhydride [of those], 3, 3', and tricarboxylic acid [, such as trimesic acid,] or acid-anhydride [of those] and acid-chloride, pyromellitic acid, 3 and 3', 4, and 4 '4, 4'-benzophenone tetracarboxylic acid or its acid anhydride is mentioned. Moreover, as an aromatic compound which has a phenolic hydroxyl group, it is benzene triol, such as benzene diols, such as phenols, such as a phenol, cresol, and a xylenol, a hydroquinone,

resorcinol, a catechol, and a methyl hydroquinone, and phloroglucine, and [0064]. A biphenol [, such as - tetramethyl biphenol,], 2, 2', and naphthols [, such as alpha-naphthol and beta-naphthol], naphthalene diol, o-phenylphenol, 2, and 2'-dihydroxy biphenyl, 2 and 2', 4, and 4'4, 4'-tetra-hydroxy benzophenone etc. is mentioned.

[0065] 0.3-1.5Eq has [the loadings of an activity ester group content aromatic compound (B)] the desirable ester equivalent to the weight per epoxy equivalent of an epoxy resin (A), and 0.5-1.0Eq 0.8-1.0Eq is used still more preferably preferably.

[0066] Preferably [since hardening of an epoxy resin is not completely performed to the weight per epoxy equivalent of an epoxy resin (A) as it is less than 0.3Eq as the ester equivalent], it is difficult to consider as the epoxy resin hardened material which has sufficiently low dielectric characteristics when 1.5Eq is exceeded, and the loadings of an activity ester group content aromatic compound (B) do not have it. [desirable]

[0067] In this invention, a reaction accelerator (C) is used with the activity ester group content aromatic compound (B) which is an epoxy resin curing agent.

[0068] As this reaction accelerator (C), the general hardening accelerator of an epoxy resin can mainly be used. As the hardening accelerator, it is organic phosphine compounds, such as imidazole compounds, such as 2-methylimidazole, 2-ethyl-4-methylimidazole, 1-benzyl-2-methylimidazole, 2-heptadecylimidazole, and 2-undecylimidazole, triphenyl phosphine, and tributyl phosphine, and [0069]. Organic phosphite compounds, such as trimethyl phosphite and triethyl phosphite, Phosphonium salt, such as ethyltriphenylphosphonium bromide and tetra-phenyl phosphonium tetraphenylborate, The salt [dicarboxylic acid / amines and DBU and terephthalic acids, such as alkylamine, such as triethylamine and tributylamine, 1, and 8 diazabicyclo (5, 4, 0)-undecene -7 (it calls for short Following DBU), / 2 and 6-naphthalene] of the origin, Tetraethyl ammonium chloride, tetrapropylammonium chloride, tetrabutyl ammonium chloride, tetrabutylammonium bromide, [0070] Quarternary ammonium salt, such as tetra-hexyl ammonium bromide and benzyl trimethylammonium chloride, 3-phenyl -1, 1-dimethyl urea, 3-(4-methylphenyl)-1, and 1-dimethyl urea, Chlorophenyl urea, 3-(4-chlorophenyl)-1, and 1-dimethyl urea, Urea compounds, such as 3-(3, 4-dichloro phenyl)-1 and 1-dimethyl urea, The salt of crown ether, such as bases, such as a sodium hydroxide and a potassium hydroxide, a potassium phenoxide, and potassium acetate, etc. is mentioned, and these can be used as independent or two or more sorts of mixture.

[0071] As loadings of a reaction accelerator (C), it is the 0.005 - 10.0 weight section to a total of 100 weight sections of an epoxy resin (A) and an activity ester group content aromatic compound (B). Under in the 0.005 weight section, if a hardening reaction is slow and exceeds the 10.0 weight sections, preservation stability will fall, and the problem that priority is given to the self-polymerization of an epoxy resin may arise.

[0072] Especially as an approach of obtaining a hardened material using the epoxy resin constituent of this invention, it is not limited but the approach of well-known common use is mentioned. For example, after dissolving in a solvent the approach and epoxy resin constituent which cast and obtain a hardened material after carrying out homogeneity mixing and carrying out heating melting of the epoxy resin constituent of this invention and considering as a varnish, it sinks into spreading and impregnation - or a glass fabric base material, solvent removal is performed with the precure of resin, and there is the approach of carrying out pressurization heating molding again etc.

[0073] If the concrete approach is mentioned, as a manufacturing method of an epoxy resin laminated sheet, a solvent is added to an activity ester group content aromatic compound (B) and a reaction accelerator (C) as an epoxy resin (A) and a curing agent, a varnish is prepared, subsequently to a glass fabric, it will sink in, it will dry for 5 minutes at 160 degrees C, and the prepreg of B stage will be manufactured. There is a method of obtaining an epoxy resin substrate or a laminate by giving metallic foils, such as gold, copper, and aluminum, to one side or both sides of this prepreg, and carrying out pressing for 1 hour on condition that a 4-8-sheet pile, 170 degrees C, and 3MPa.

[0074] The various organic solvents which are dissolved in homogeneity and deal in an epoxy resin, an activity ester group content aromatic compound, and a reaction accelerator as [both] a solvent in which

the epoxy resin constituent of this invention is dissolved are used. Although it cannot generally ***** according to the class of epoxy resin to be used, generally mono-ether glycol system solvents, such as aromatic hydrocarbon system solvents, such as ketone solvent, such as amide system solvents, such as - dimethylformamide, and N,N, N'-dimethylacetamide, an acetone, a methyl ethyl ketone, methyl isobutyl ketone, and a cyclohexanone, toluene, and a xylene, the ethylene glucol monomethyl ether, and ethylene glycol monobutyl ether, etc. are mentioned. [N-methyl pyrrolidone, N-methyl formaldehyde, N, and] These solvents may be used as one sort or two sorts or more of mixture.

[0075] Usually, at the reaction of an epoxy resin and a curing agent, a polar high hydroxyl group generates in connection with the ring breakage of an epoxy group in many cases. For example, the curing agent of an amine system or a phenol system gives a hydroxyl group, and an acid-anhydride system curing agent becomes a carboxylic acid easily by moisture absorption, and it gives a hydroxyl group as a result. However, if an activity ester group content aromatic compound is used as a curing agent of an epoxy resin, with the ring breakage of an epoxy group, this activity ester group will react, a polar low ester group will generate, and a hydroxyl group will not be generated. That is, while the water absorption property of an epoxy resin hardened material becomes good, it becomes a low dielectric, i.e., a low dielectric constant, and a low dielectric dissipation factor. Moreover, when this activity ester group content all aromatic compound is used, since many rings are included, high thermal resistance and fire retardancy are shown.

[0076] Since the epoxy resin constituent of this invention can obtain the hardened material which was excellent in dielectric characteristics, especially a dielectric dissipation factor compared with the conventional epoxy resin, the obtained low dielectric ingredient

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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

CLAIMS

[Claim(s)]

[Claim 1] The epoxy resin (A) which has two or more epoxy groups in 1 molecule, the aromatic compound (B) which has two or more activity ester groups in 1 molecule obtained from the compound which has a multiple-valued carboxylic acid, and the aromatic compound which has a phenolic hydroxyl group, the epoxy resin constituent for low dielectric ingredients which uses a reaction accelerator (C) as an indispensable component.

[Claim 2] The epoxy resin constituent for low dielectric ingredients according to claim 1 with which a low dielectric is characterized by being 3.5 or less dielectric constant.

[Claim 3] The epoxy resin constituent for low dielectric ingredients according to claim 1 or 2 with which a low dielectric is characterized by being 0.01 or less dielectric dissipation factor.

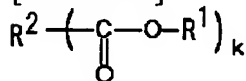
[Claim 4] The activity ester group content aromatic compound (B) to the epoxy resin (A) which has two or more epoxy groups in 1 molecule comparatively It is 0.3-1.5Eq in the ester equivalent of this aromatic compound (B) to the weight per epoxy equivalent of this epoxy resin (A). The epoxy resin constituent for low dielectric ingredients given in any 1 term of claims 1-3 to which a reaction accelerator (C) is characterized by being the 0.005 - 10.0 weight section to a total of 100 weight sections of an epoxy resin (A) and an activity ester group content aromatic compound (B).

[Claim 5] The epoxy resin constituent for low dielectric ingredients given in any 1 term of claims 1-4 characterized by being all the aromatic compounds (B') with which the aromatic compound (B) which has two or more activity ester groups in 1 molecule obtained from the compound which has a multiple-valued carboxylic acid, and the aromatic compound which has a phenolic hydroxyl group has two or more activity ester groups in 1 molecule obtained from the aromatic compound which has a multiple-valued carboxylic acid, and the aromatic compound which has a phenolic hydroxyl group.

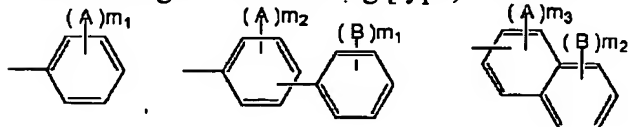
[Claim 6] The epoxy resin constituent for low dielectric ingredients given in any 1 term of claims 1-4 characterized by the aromatic compounds (B) which have two or more activity ester groups in 1 molecule obtained from the compound which has a multiple-valued carboxylic acid, and the aromatic compound which has a phenolic hydroxyl group being all aromatic compounds expressed with a general formula (1).

General formula (1)

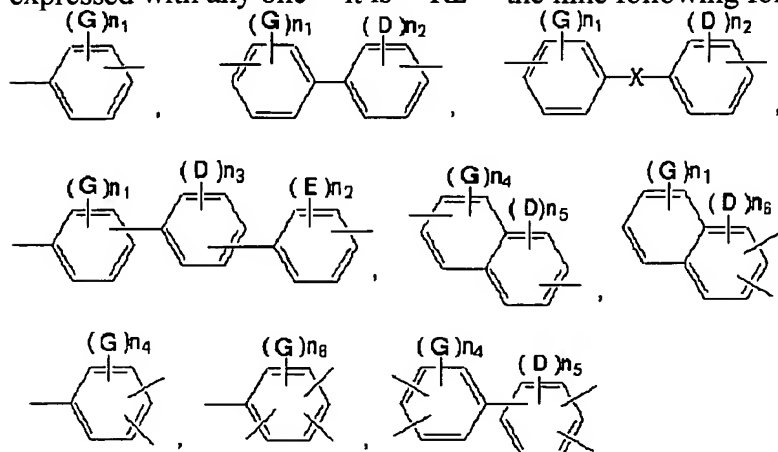
[Formula 1]



k is the integer of 2-4 among [type, and R1 is the three following formulas [** 2].



(-- the inside of a formula, and A and B -- each -- the same -- or it differs, a halogen atom and an alkyl group are shown, and, as for 0-5, and m2, m1 shows 0-3, as for 0-4, and m3.) -- the aromatic series ring expressed with any one -- it is -- R2 -- the nine following formulas [** 3]



(-- the inside of a formula, and D, E and G -- each -- independently the same -- or -- differing -- a halogen atom and an alkyl group -- being shown -- X -- a sulfur atom, an oxygen atom, SO₂, or CO -- being shown -- n1, n2, and n3 -- each -- independent -- 0-4, and n4 and n5 -- each -- 0-3, and n6 show 0-2 independently.) -- it is the aromatic series ring expressed with any one.]

[Claim 7] The epoxy resin constituent for low dielectric ingredients given in any 1 term of claims 1-6 characterized by the weight per epoxy equivalent of the epoxy resin (A) which has two or more epoxy groups in 1 molecule being 100-1000.

[Claim 8] The epoxy resin constituent for low dielectric ingredients given in any 1 term of claims 1-7 characterized by reaction accelerators (C) being an imidazole compound, an organic phosphine compound, an organic phosphite compound, phosphonium salt, amines and its salt, a urea compound, bases, and one or more sorts of mixture chosen from the group which it becomes from the salt of crown ether.

[Claim 9] The low dielectric ingredient which hardens the epoxy resin constituent for low dielectric ingredients of a publication in any 1 term of claims 1-8, and is obtained.

[Claim 10] The low dielectric substrate characterized by having a metallic foil to one side or both sides of the film which consists of a low dielectric ingredient according to claim 9, or a sheet.

[Translation done.]